ULTRASONIC ABSORPTION AND RELAXATION MECHANISM

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ABSTRACT. It is considered that absorption of elastic waves is always due to some sort of relaxation mechanism which may be of varied nature. The general relationship to express the co-efficient of absorption may, however, be put in the same form for different types of relaxation mechanism, with two constants which are determinable by the process of relaxation mechanism. As such, the absorption due to viscosity considered by Stokes, is also regarded as arising from a relaxation mechanism termed as frictional relaxation mechanism. On this basis the usual Stokes relation for absorption co-efficient requires modification. From a systematic correlation between the discrepancy in experimental and calculated values of α/ν^2 on Stokes theory on the one hand and the broadening of elastic wave diffraction spectral lines on the other hand, for different liquids, it has been argued that the same relaxation mechanism should explain the discrepancy in different cases and this has been considered to be due to the necessary modification of the Stokes theory of absorption. Thus, it is considered that the absorption of elastic waves is mainly due to the frictional relaxation mechanism in different liquids.

The phenomenon of absorption of elastic waves, due to various causes can always be considered to be caused by a particular relaxation mechanism. In accordance with the different causes the relaxation mechanism would evidently be different. These are, however, always expressible in the general form,

$$\mu/V\nu=\alpha/\nu^2=\frac{A_1}{V.\nu_m}\cdot\frac{1}{1+(\nu/\nu_m)^2}\equiv\frac{A}{\nu_m}\cdot\frac{1}{1+(\nu/\nu_m)^2} \qquad \ldots\ (1)$$

In the particular region, where $\nu<<\nu_m$, one may write the absorption relation in the form,

$$\alpha/\nu^2=A/\nu_m=\text{const.} \qquad \ldots\ (2)$$

Here, μ and α are the absorption co-efficients per unit wavelength and per unit distance, the constant A is a function of the wave velocity V, and may be considered as a measure of the absorption energy due to the particular cause considered. The quantity $1/\nu_m$ is a measure of the relaxation time or the time required by a particle for the transfer of energy from the initial condition to the changed condition. In most of the cases, the constant A,

to some extent, and the relaxation time $1/\nu_m$, are unknown quantities and require to be found by the experimental results. There appears, however, the apparent difficulty that when there are more than one plausible cause of relaxation, we do not know why we should choose one particular mechanism in preference to others, to explain the absorption. The difficulty, nevertheless, disappears when the constants are determinable.

The experimental results on the absorption of ultrasonic waves in liquids have been summarised in a paper by Pinkerton (1949). They are, in many cases, much larger than the absorption calculated on Stokes' theory (Stokes, 1845). It was suggested initially by Dutta and Ghose (1937-38) and also by Kneser (1938), that the excess absorption from Stokes' theory, in the case of liquids, might be due to a relaxation process associated with excitation to vibrational states.

The difficulties, with the excitation relaxation mechanism to explain the excess, have been many. It was pointed out (Dutta and Ghose, 1937-38 ; Herzfeld, 1941) that the irregular absorption in water could not be explained by the excitation to higher vibrational states, as the measure of absorption due to vibrational excitation becomes much too small in this case, to make any appreciable contribution to absorption.

A different process of relaxation mechanism, as that due to the changes in the molecular arrangements of the minutest particles, was put forth (Hall, 1947) to explain these cases. We are thus presented with an excitation relaxation mechanism to account for the excess absorption in non-associated molecules and a rearrangement relaxation mechanism to account for the excess of absorption in associated molecules. To fit in with the experimental results one has only to give suitable values to A and ν_m.

In a recent note (Lamb and Andreae, 1951), attempt has been made to establish positively, the excitation relaxation mechanism as the cause of excess absorption, in the case of carbonbisulphide. To conform to the experimental results, they have obtained the values of A_1 as 0.46 and ν_m as 72 megacycles per second. The amount of unaccounted for absorption on this basis, has been calculated by them to be given by $\alpha/\nu^2 = 428 \times 10^{-17}$. This is much too large compared to the other main contributory part of absorption, namely, the Stokes' absorption, which gives a value of 5×10^{-17} units only. We are, thus, again confronted with an excess absorption which has to be explained anew.

Further, from a study of the excitation states in different molecules it has been considered by various workers (Pinkerton, 1949 ; Dutta and Ghose, 1937-38 ; Fricke, 1941) that the constant A_1 would be of the order of 0.25 for benzene. To explain the experimental value of excess α/ν^2, this gives, on the basis of relaxation mechanism, the ν_m value for benzene as 3×10^8 cycles per second. Thus at an impressed frequency of the same

value, the measure of absorption co-efficient in benzene would drop down from the α/ν^2 value of 800×10^{-17} to a value of 400×10^{-17}. The experimental results of Rapuano (1947) do not give this indication. A large variation of the ν_m values of benzene and carbon bisulphide is, also, not understandable.

Besides, the experimental results of Pinkerton (1948) on acetic acid, so far as the downward trend of α/ν^2 is concerned, are exactly of a similar type to the results of Rapuano (1950) on carbon bisulphide. There is only a shift in the frequency scale. No process of excitation mechanism would, however, explain the large excess absorption in the case of acetic acid, as this would require much too large value of A_1 or much too small value of ν_m.

It, thus appears that the excitation relaxation mechanism is without positive support from any case of absorption in liquids. Even when we try the mechanism in the case of CS_2, we have to seek for another relaxation process to explain the absorption left over, and this is difficult to conjecture. We have, thus, to leave out the relaxation mechanism due to excitation as a major contributory cause of absorpion.

From the consideration of the close relationship between the diffuseness and breadth of the lines of elastic wave diffraction spectra (Dutta, 1952), indicative of a range of velocity, and the departure of the α/ν^2 values from the Stokes' values, irrespective of the amount of absorption and of the associated or the unassociated states of the medium, it requires to be considered that all the liquids have to be treated by an identical relaxation mechanism. For, the departure from the Stokes' absorption value has shown a uniformly varying property of the molecules in the form of spectral line widths, and this must have been impressed by any particular relaxation process.

It may be considered, that the Stokes' absorption relation,

$$\alpha/\nu^2 = 8\pi^2\eta/3\rho V^3 \qquad \ldots \quad (3)$$

also corresponds to a particular relaxtion mechanism, namely, the frictional relaxation mechanism. Thus, the absorption should have been properly represented by a relation of the type of equation (1). The constant A would have been a measure of the absorption of energy, due to the overcoming of the frictional force and ν_m would have been a measure of the time required for the transfer of energy from the initial condition to the condition of motion after overcoming the frictional force. In the usual Stokes' relation, the frequency ν has been considered to be much too small compared to the characteristic constant ν_m, and thus, the second factor has not occurred in the Stokes' relation. On the basis of a justifiable, variable force of friction and restitution, however, acting on the molecules, the complete ultrasonic absorption data may be explained on the basis of a frictional relaxation mechanism, with the help of the necessary and implied modifications of the Stokes' theory. This has been shown in a recent paper (Dutta, 1952).

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